CO Preferential Oxidation Supports Study for Fuel Cell Applications

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DOE NETL UCR/HBCU Contractors Review Meeting
June 3rd, 2003

Current Address:
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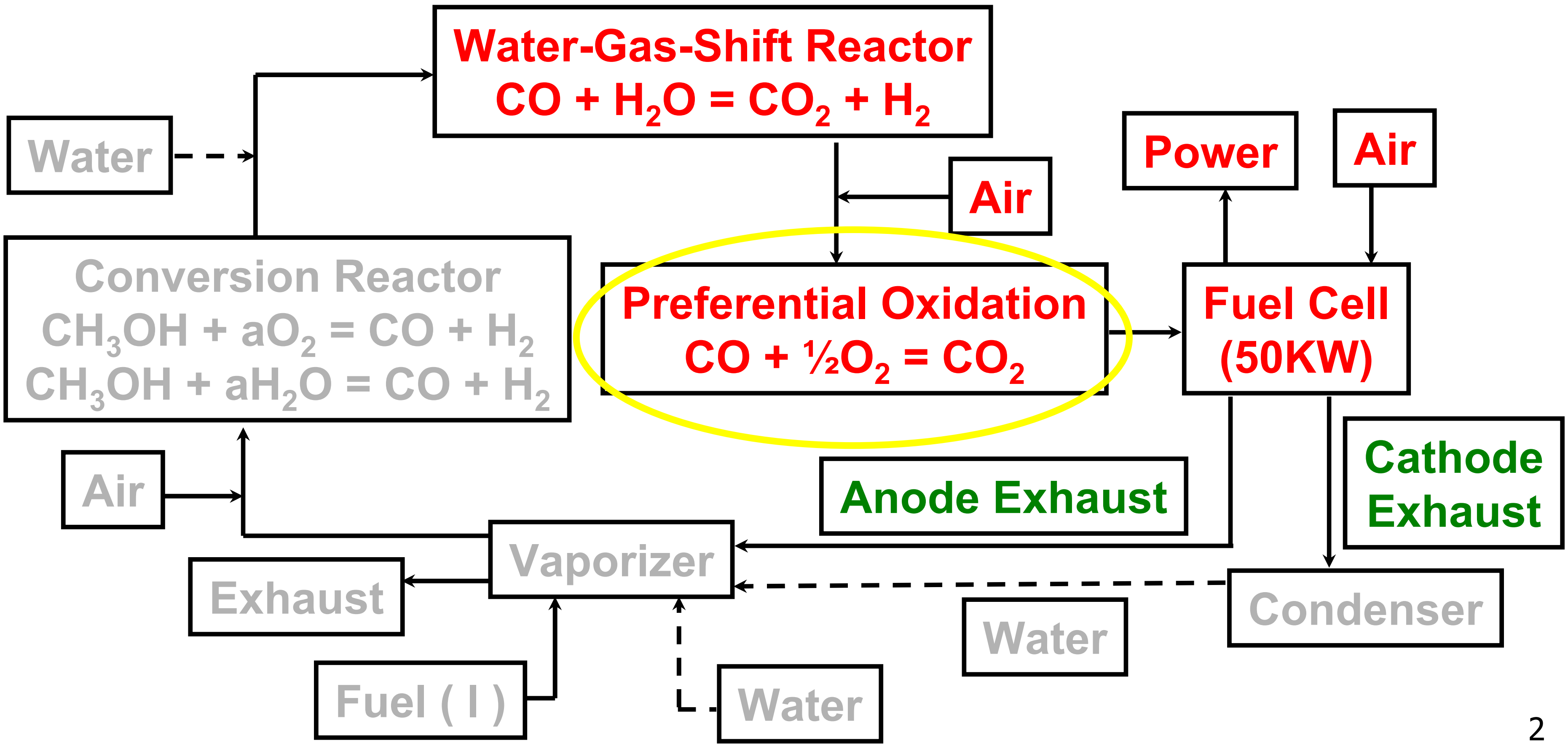
PEM Fuel Cell

Fuel Source - Reformulated Coal Derived Fuels:

Methanol, Natural Gas, or Gasoline.



Preferential Oxidation (PROX)

- Objective
  - Reduce CO from 1% to < 10 ppm to eliminate CO poisoning of the anode in proton exchange membrane (PEM) fuel cells.

- Reactions

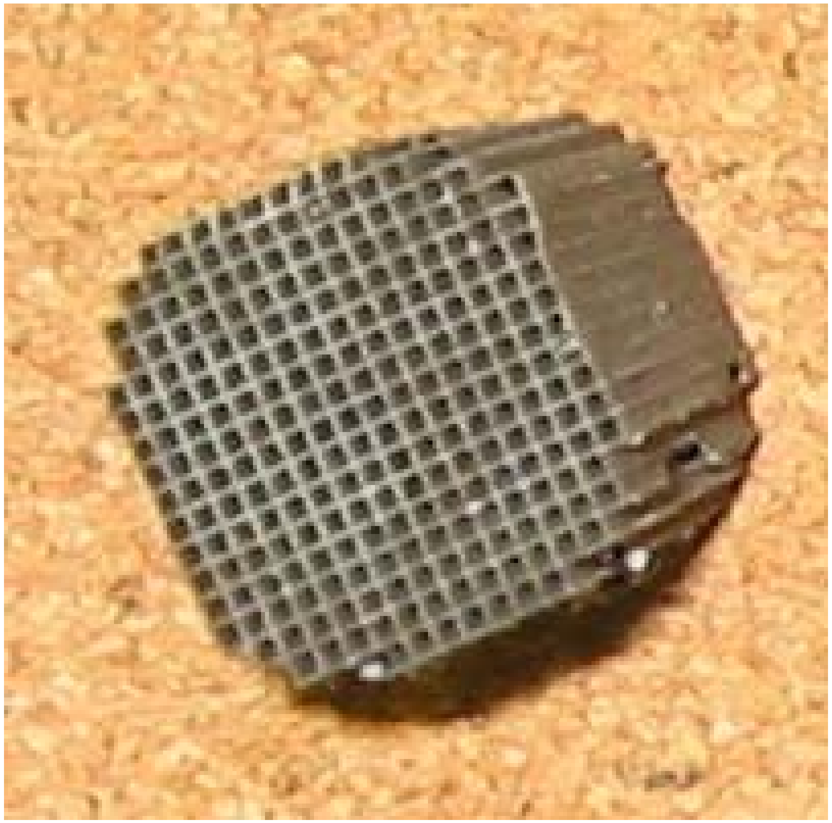
$$CO + \tfrac{1}{2}O_2 \longrightarrow CO_2$$

$$H_2 + \tfrac{1}{2}O_2 \cancel{\longrightarrow} H_2O$$

$$(CO_2 + H_2 \longleftrightarrow CO + H_2O)$$

- Requirements
  - High CO Conversion (99.9%)
  - High CO Selectivity

Straight-Channel Monoliths



Ceramic
(400 cells per square inch)

Metal
(400 cells per square inch)

Metal Foam Monoliths

- High thermal conductivity
- Reticulated structures, high surface area
- Lateral (radial) mixing
- Low density, high strength structure

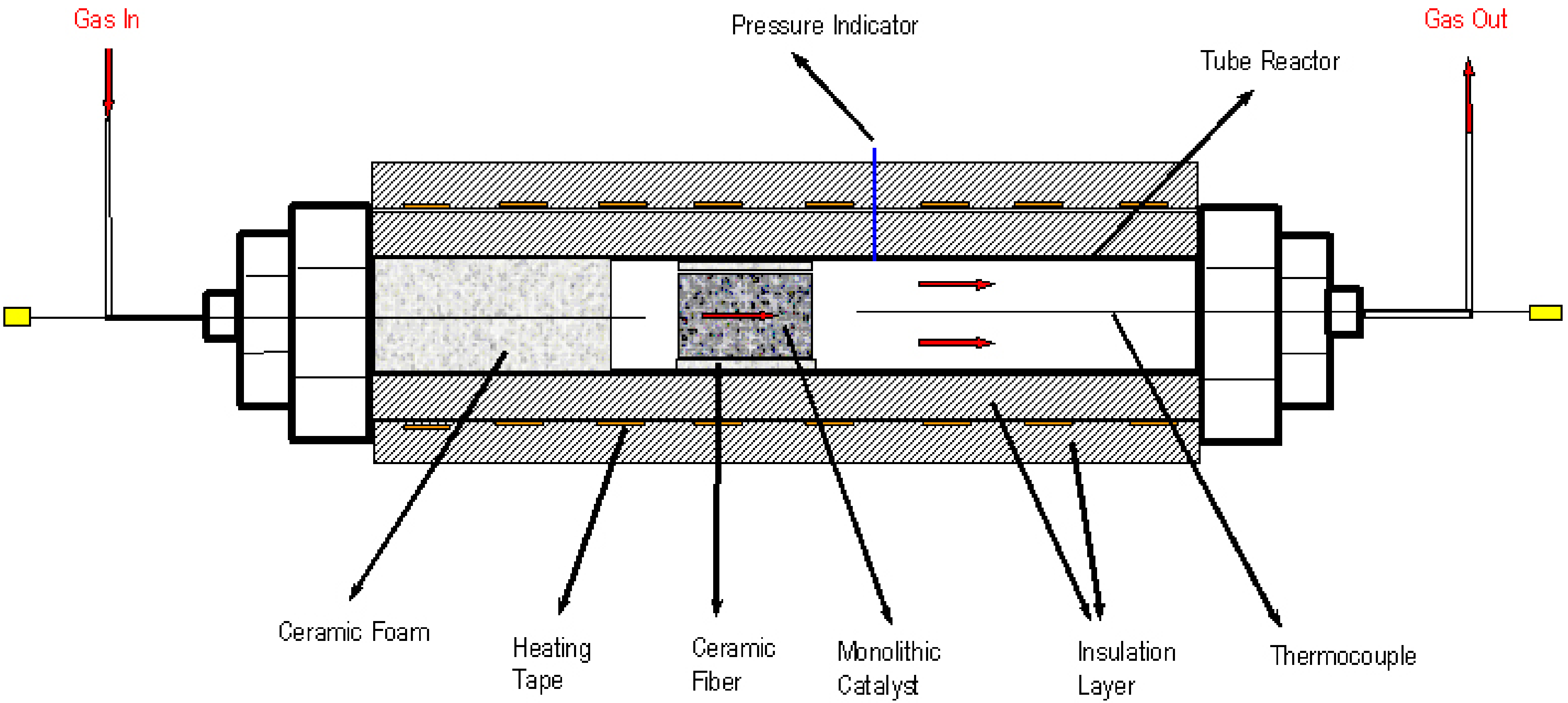
Effects Studied

I. Catalyst Composition
- 5% Pt; Varying Fe Loading*
  (Washcoat loading ~ 1.6 g/in^3)
- Support Structure

II. Operating Variables
- Inlet Temperature
- Inlet CO Concentration
- Inlet O_2 Concentration
- Space Velocity
- Linear Velocity

* Straschil, H. K.; Egbert, W., Jr., Canadian Patent 828058 (11/25/69).
Korotkikh, O.; Farrauto, R., Catalysis Today, 62 (2000), 249-254.
Liu, X.; Korotkikh, O.; Farrauto, R., Applied Catalysis A: General, 226 (2002) 293-303.

Experimental Reactor



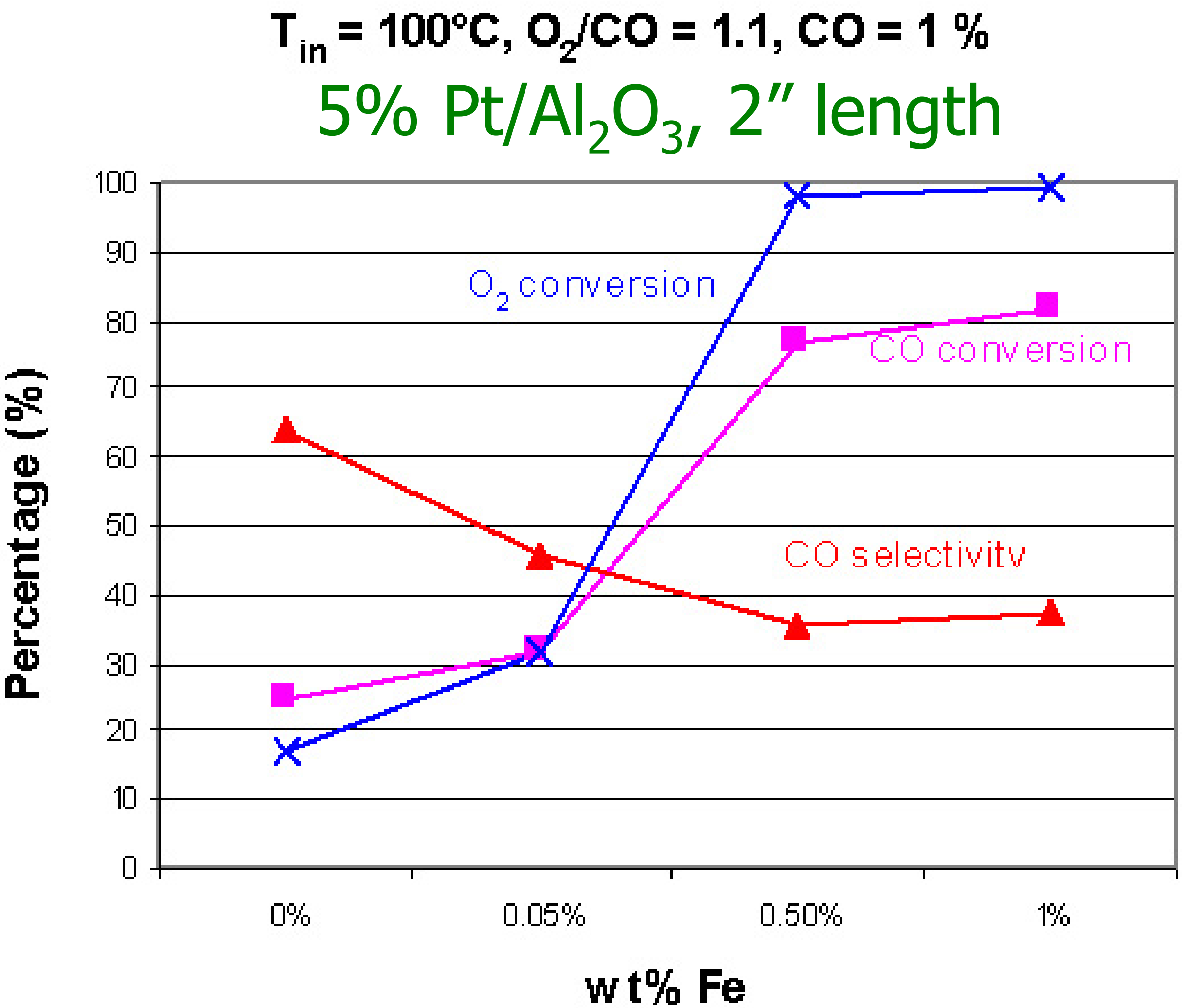
Close to Adiabatic

Range of Test Conditions

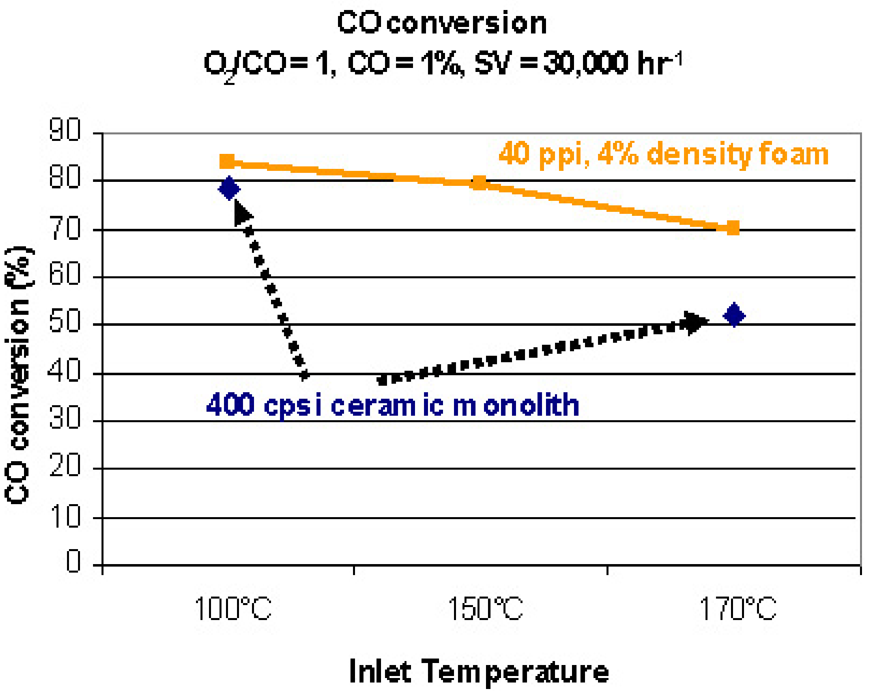
- Catalyst Length: 2 – 6 in.
- Pressure: 2 atm (abs.)
- Space Velocity: 5,000 - 60,000 hr^{-1}
- CO Inlet Concentration: 0.1% - 1.0%
- O_2/CO ratio: 0.25 - 1.0
- Inlet Temperature: 80°C - 170°C
- Inlet Gas Composition:

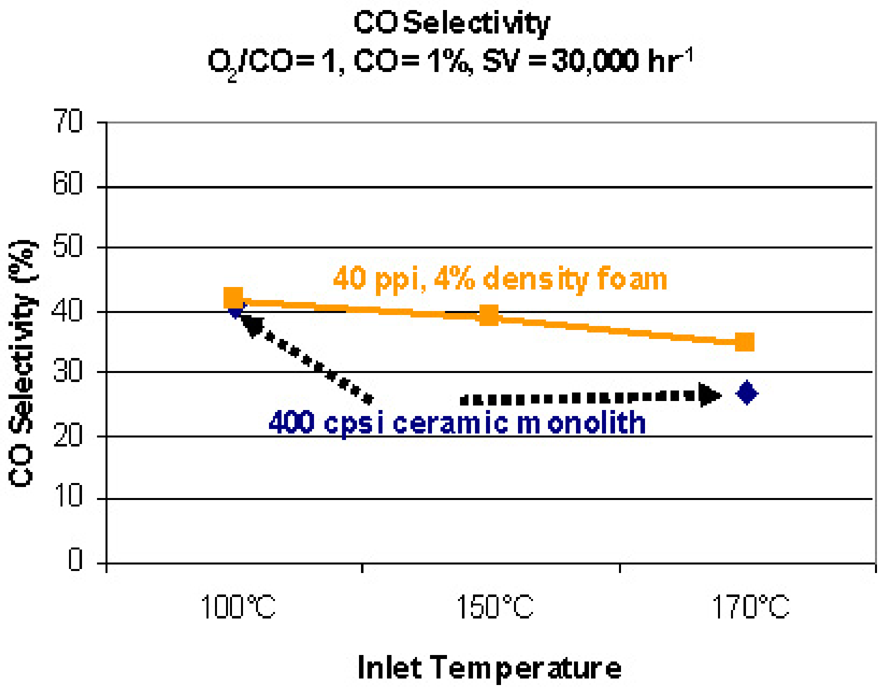
H_2	CO_2	H_2O	CO	O_2	N_2
42%	9%	12%	X%	Y%	Balance

Fe Loading on Ceramic Monolith



Support Comparison

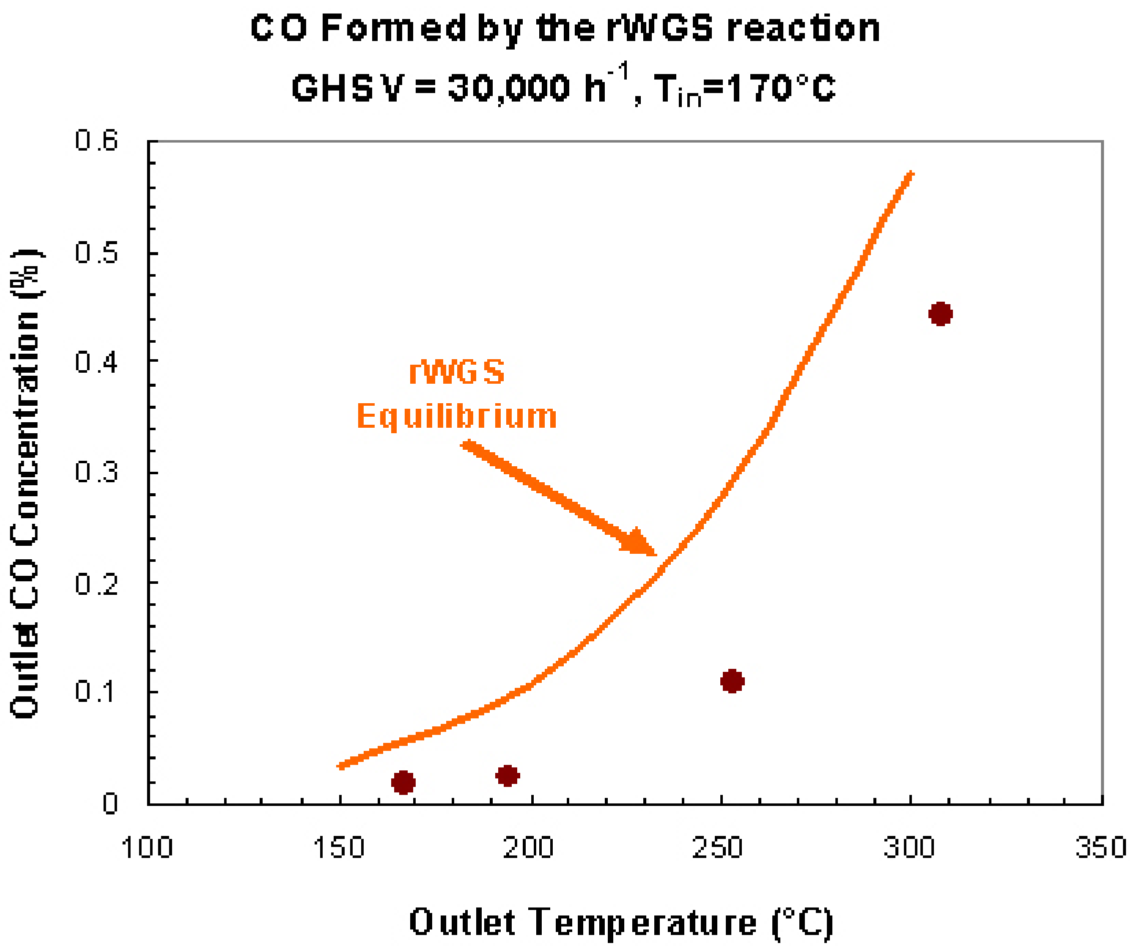




5% Pt/0.5% Fe; 2” Supports

rWGS Reaction

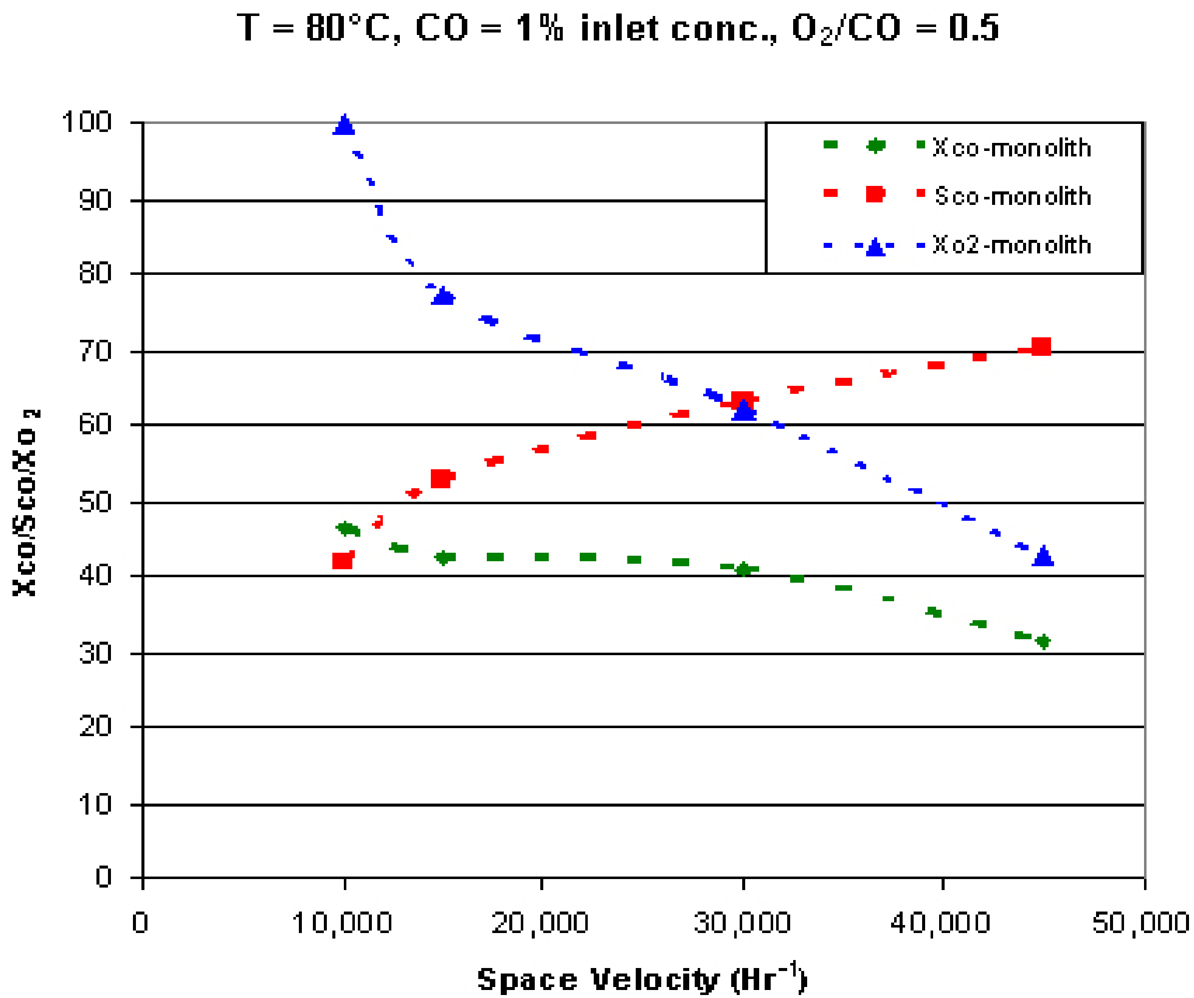
No CO in Feed, Varying O_2 in Feed



Equilibrium assumes all O_2 reacted

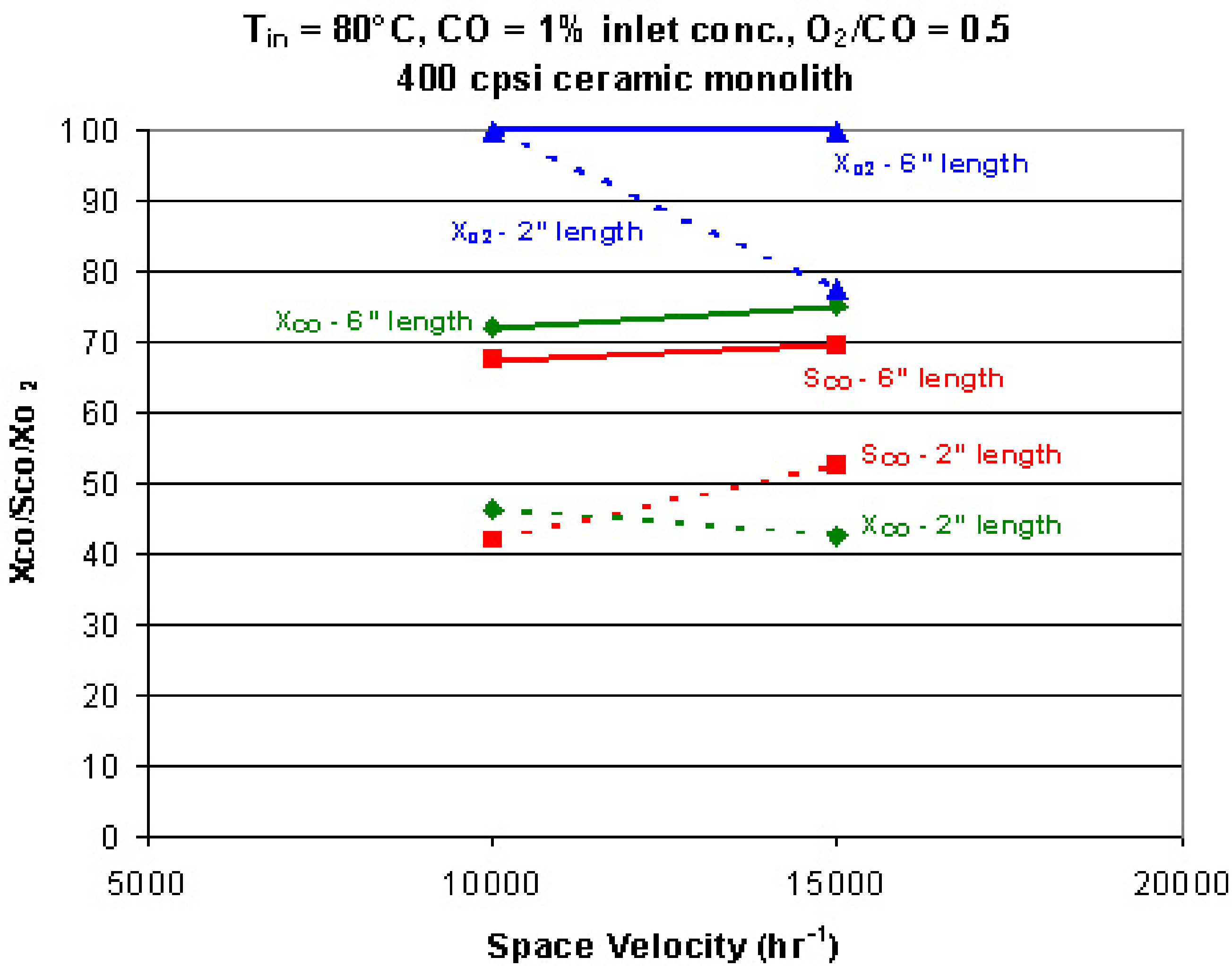
5% Pt/0.5% Fe; 2" Ceramic Monolith

Effect of Space Velocity



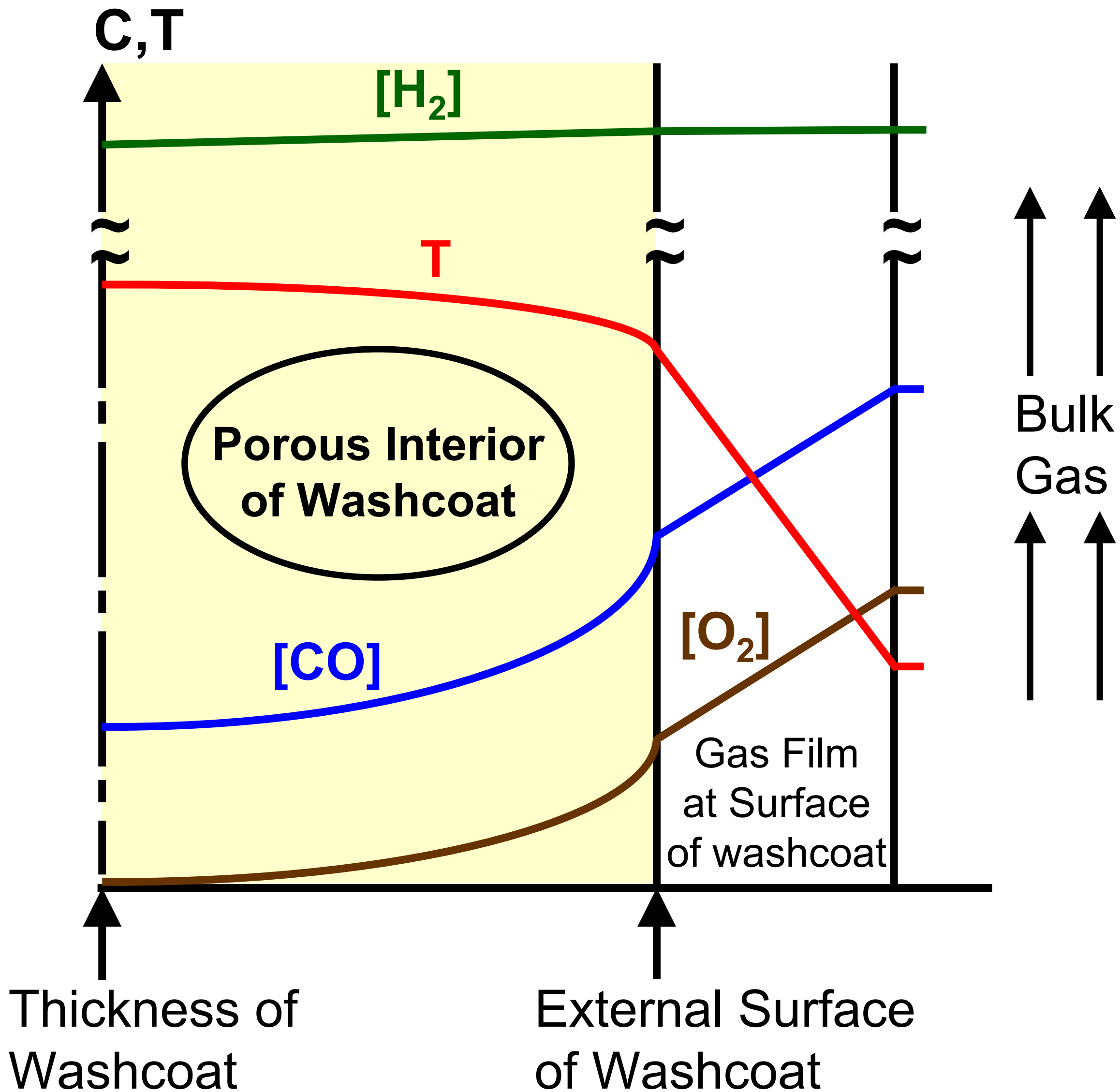
5% Pt/0.5% Fe; 2" Ceramic Monolith

Effect of Linear Velocity

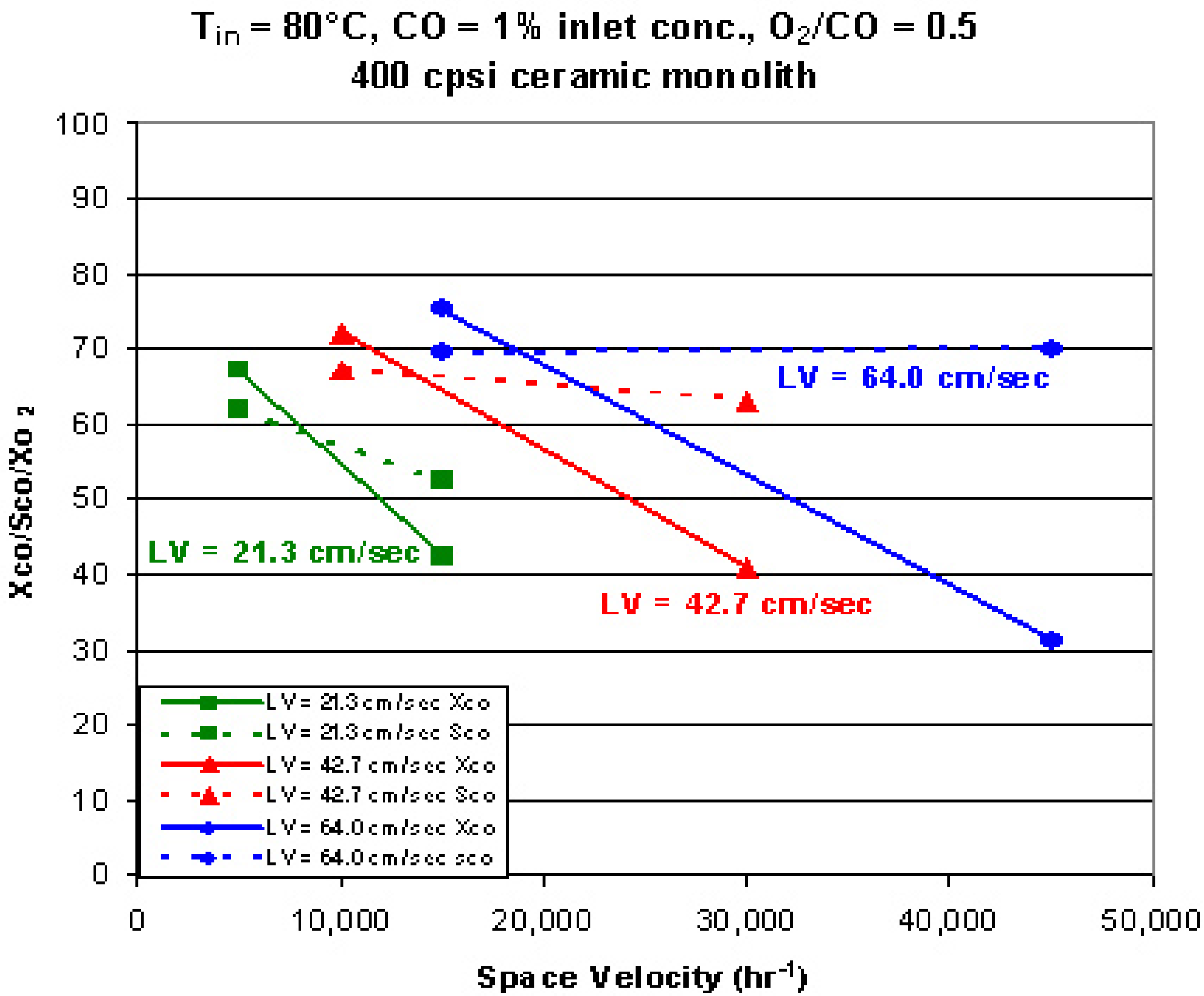


5% Pt/0.5% Fe; 2" & 6" Ceramic Monolith

Potential Transport Effects in PROX

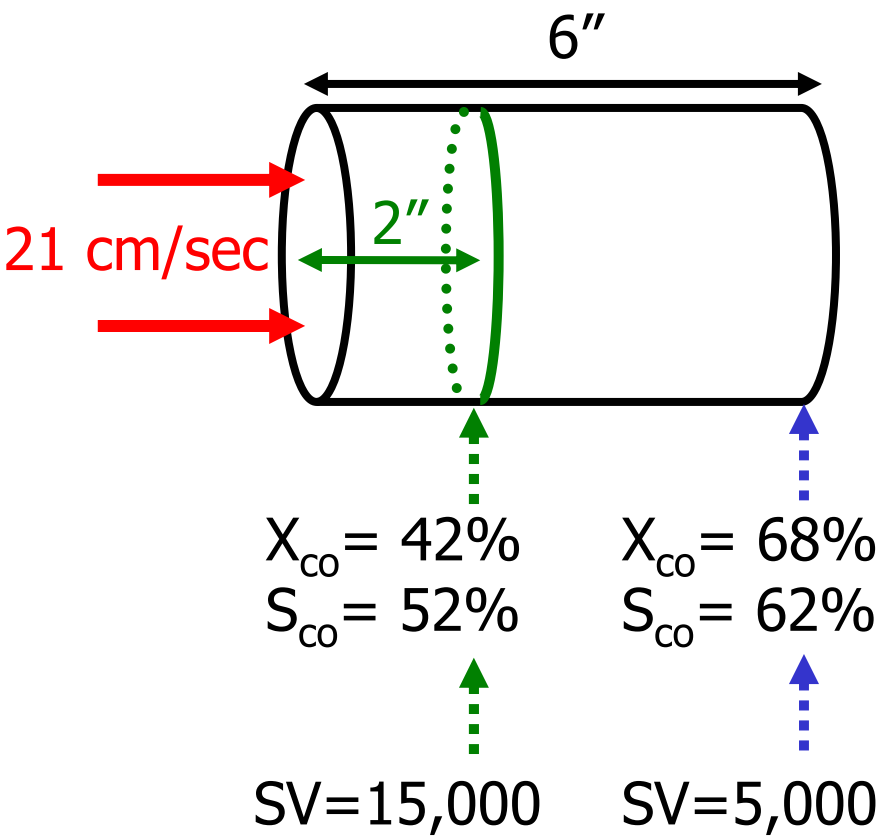


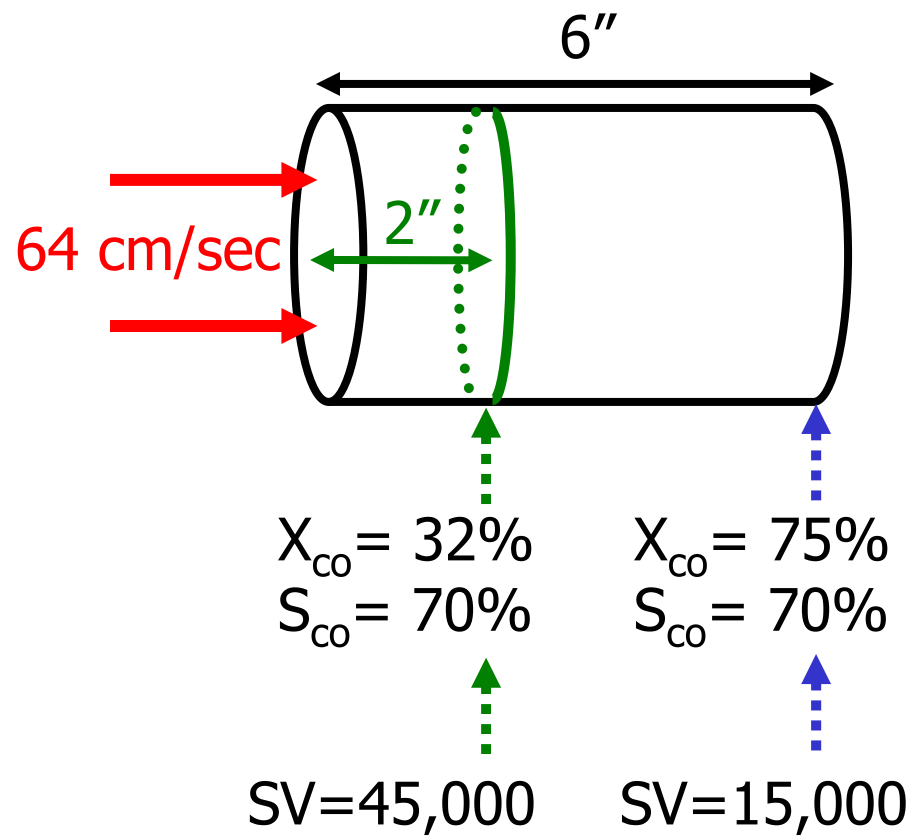
Effect of Space Velocity



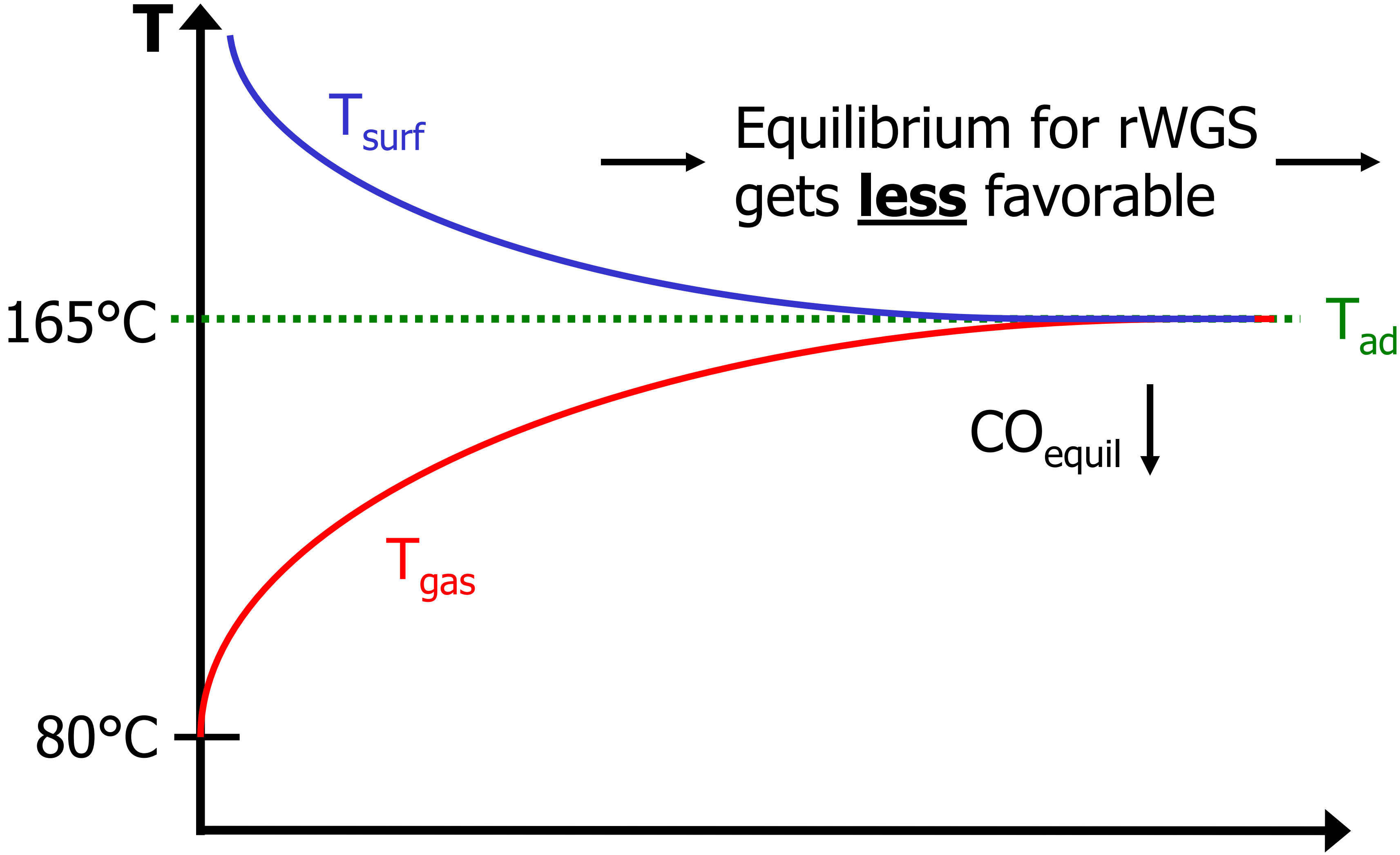
5% Pt/0.5% Fe; 2” & 6” Ceramic Monolith

Effect of Linear & Space Velocity





Temperature Profiles in Catalyst



Conclusions

- rWGS reaction can limit CO conversion.
  - Need low T (or catalyst with no rWGS activity)
  - Need to avoid transport influence
- External heat/mass transport can be important.
- Support must have:
  - High external surface area/volume
  - High heat/mass transfer coefficients
- Straight-channel monoliths may not be best.

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  - Ken Butcher
  - Anil Prabhu

- Kyle Bishop, University of Virginia

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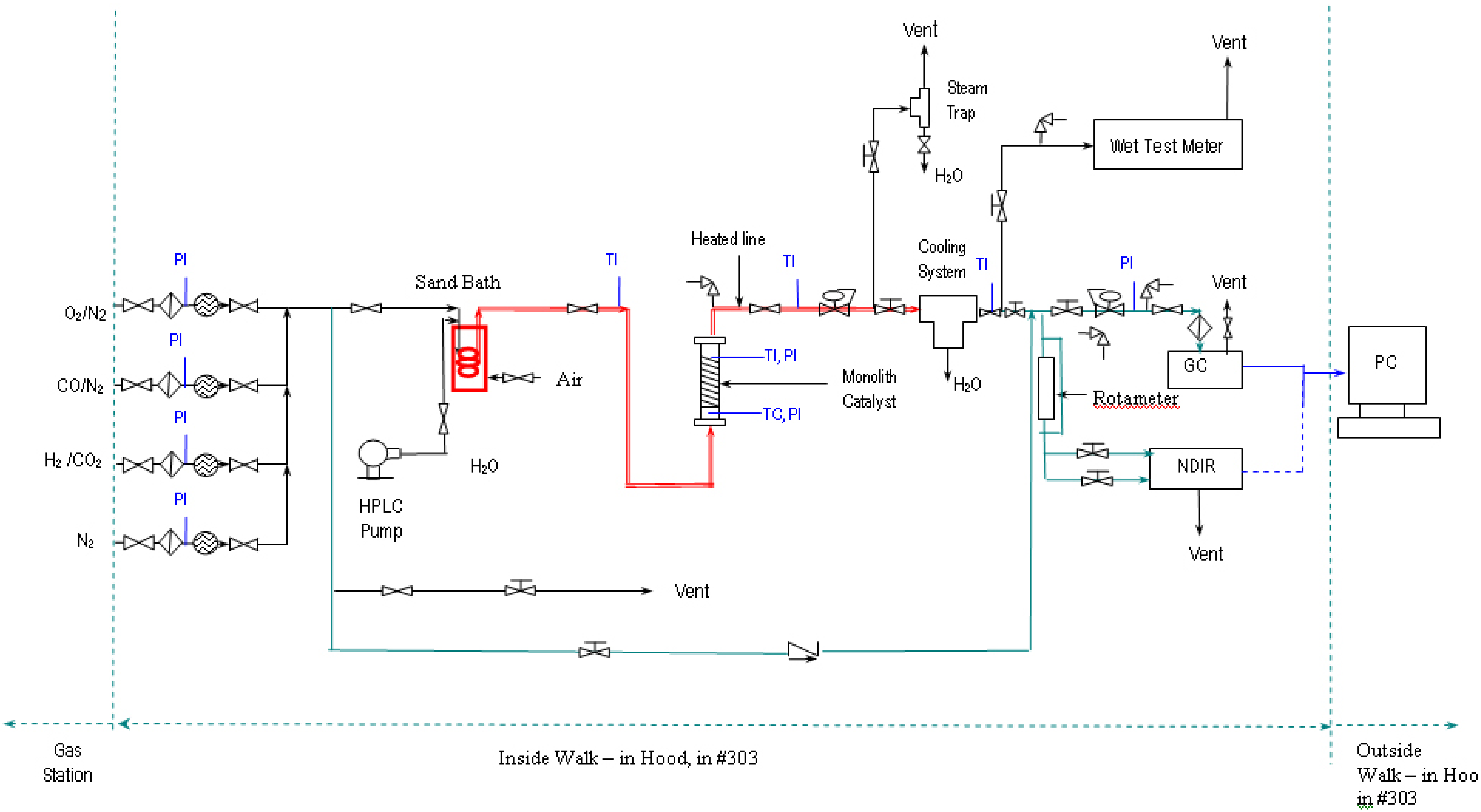
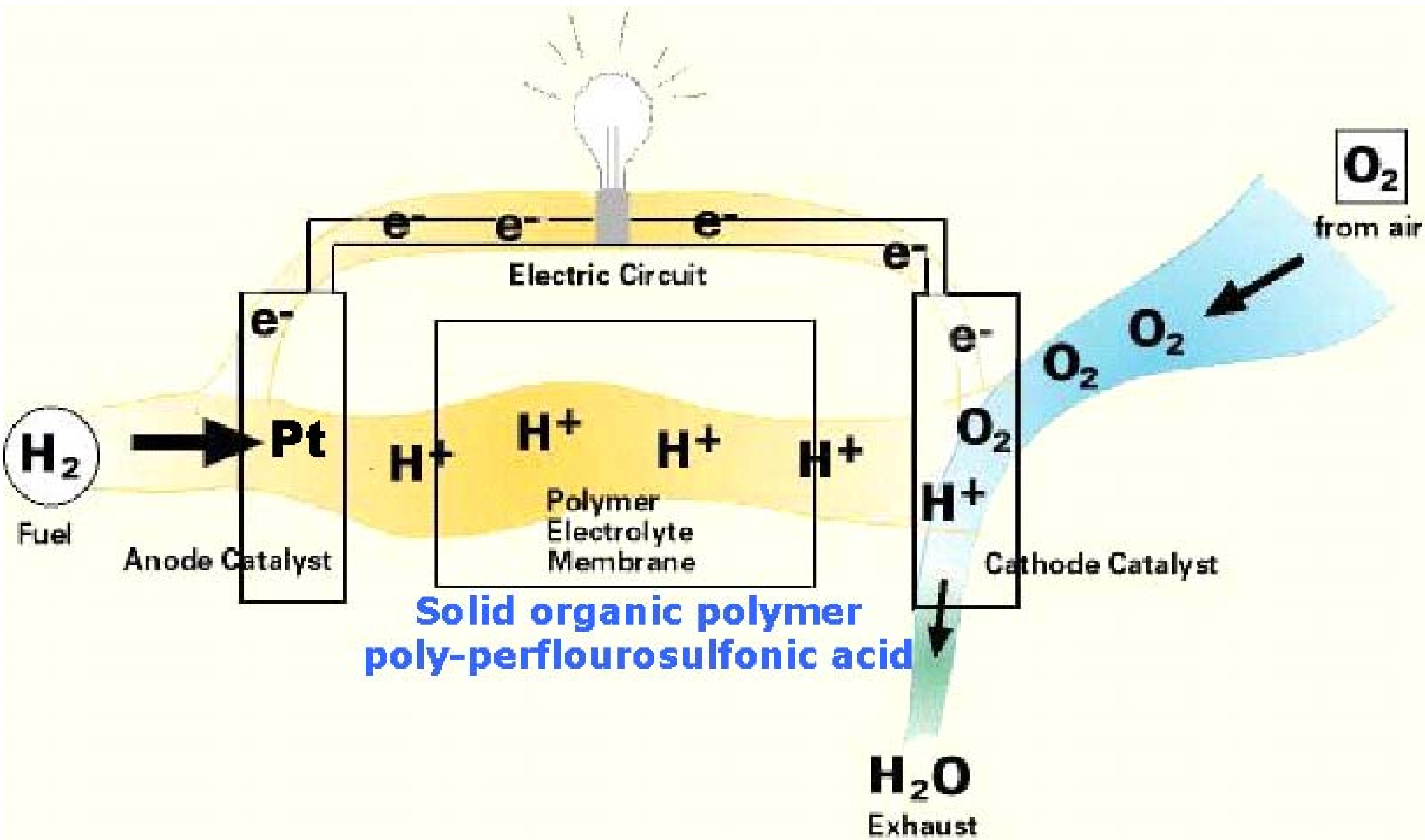
Process Design

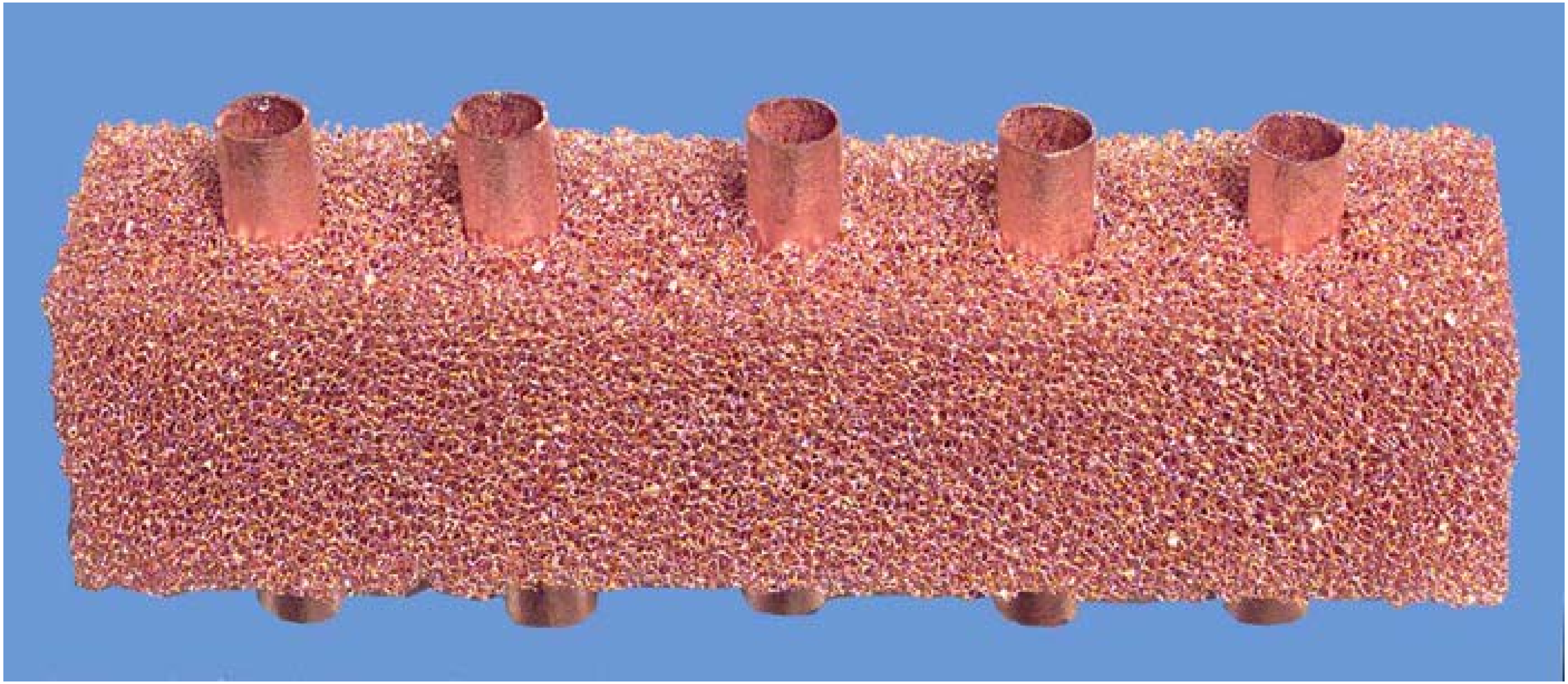


PEM Fuel Cell



* From http://www.fuelcells.org

Foams with Tubes



Catalyst Preparation

1. Dip the catalyst support in a slurry containing Al_2O_3. Blow off excess slurry with compressed air. Dry the washcoated support at 100^oC and dip again as necessary.
2. Calcine at 500^oC to insure washcoat adhesion.
3. Determine the pore volume of the washcoat by impregnating the support with deionized water.
4. Impregnate the washcoated support with a Pt solution.
5. Dry at 100^oC, then calcine at 500^oC
6. Impregnate the supported catalyst with a Fe solution.
7. Dry at 100^oC, then calcine at 500^oC